

# MEDAL: A Time-Dependent Computational Methodology for Very High Burnup Systems

W. C. Sailor and J. W. Davidson  
Los Alamos National Laboratory  
Los Alamos, NM 87545 USA

**Abstract:** A calculational methodology which couples the transport code MCNP4a with the depletion code ORIGEN2.1 using a modified Euler method is described in this paper. The method allows for depletion calculations for very high burnup systems where there are large spectral changes with time and is more accurate than the simple Euler method. Time-dependent calculational results of the evolution of the actinide species in a weapon plutonium-fueled, molten salt system are given as an example of the utility of the procedure.

## Introduction

In order to quantify the burnup of weapon plutonium in high-flux thermal systems it is necessary to perform depletion calculations under conditions where the neutron energy spectrum in the system is strongly time-dependent. This is in contrast to a hypothetical system where, if there were no spectral changes with time, the equations governing the buildup, consumption and decay of radionuclides would be linear.

The depletion analysis code ORIGEN2.1<sup>1</sup> is an example of a code that is exact for such a linear system. In general, the code solves N coupled equations governing the rate at which the amount of nuclide i changes as a function of time. One such equation is described as:

$$(1) \quad \frac{dX_i}{dt} = \sum_{k=1}^N L_{ik} \lambda_k X_k + \phi \sum_{k=1}^N f_{ik} \sigma_k X_k - (\lambda_i + \phi \sigma_i + r_i) X_i + F_i$$

where  $X_i$  is the atom density of nuclide i, N is the number of nuclides in the system,  $L_{ij}$  is the branching ratio for radioactive decay from species j to species i,  $\lambda_i$  is the radioactive decay constant,  $\phi$  is the position and energy-averaged neutron flux,  $f_{ik}$  is the branching ratio for neutron capture reactions that lead from species k to i,  $\sigma_k$  is the spectrum-averaged neutron absorption cross section of nuclide k, and  $F_i$  is the feed rate of the species i. If all the coefficients on the right-hand-side of (1) are independent of all the  $X_i$ , the equations are linear. Also, if a system operates at a constant specific power, and all cross section ratios were constant, the system would remain linear. However, cross section ratios in general change with time. This is especially true in systems absent a resonance absorber. The dependence of average cross section on concentration is

unique to each system and must be found through some explicit neutron transport calculation. One code performs a fully-coupled depletion/transport calculation, but it has only one spatial dimension and uses only a fixed 26-group structure.<sup>2</sup> At present, there are no three-dimensional transport codes that also perform fully coupled depletion calculations.

In order to solve this problem in approximate fashion, we have coupled together the codes MCNP4a<sup>3</sup> and ORIGEN2.1 in a self-consistent fashion in a scheme called the Modified Euler Depletion Analysis Loop (MEDAL). The code MCNP4a is a three-dimensional Monte Carlo neutron and photon transport code that uses continuous energy-dependent cross section sets, from the Evaluated Nuclear Data File (ENDF/B-V)<sup>4</sup> or the Evaluated Nuclear Data Library (ENDL)<sup>5</sup>. The unmodified "Euler method" refers to a method for solving differential equations by evaluating any non-linear coefficients at the beginning of a time step only, then using an explicit forward time step.<sup>6</sup> Here, it would mean coupling MCNP4a and ORIGEN2.1 together in a simple 1 - 2 - 1 - 2 . . . repetitive sequence, as is done in the code package MOCUP.<sup>7</sup> In **Figure 1** we show a computational scheme of our own that is similar to this package. Under the Modified Euler method, as shown in **Figure 2**, the one-group cross sections are first evaluated at the beginning of a time step. The code KOLAPS writes these cross sections to a library that is used by ORIGEN2.1 to perform a *temporary* forward time step calculation. The inventory is extracted with the code TDI from the ORIGEN2.1 output file, and then is written to an MCNP4a input file. The spectrum is calculated again, giving another set of one-group cross sections. At this point in the calculation, the code KOLAPS reads both these MCNP4a results and the results at the beginning of the time step and averages them together. These average values are used to perform the forward time step that is actually saved and used by the routine. The temporary forward-time-step results are discarded. It is believed that this procedure is more reliable under more circumstances than an unmodified Euler method per unit of computer time used.

In this paper unmodified and modified Euler approximations to model equations that have known analytical solutions are first presented. Computational comparisons are then presented in this paper for the methods shown in Figures 1 and 2 as applied to a high-flux weapon-Pu burning system.

### Simplified Depletion Model

In order to compare the unmodified Euler method with a modified Euler method in the case of a coupled transport/depletion case, we have used a simplified model of a liquid-fuel plutonium-burning system. This model corresponds as closely to the systems that we are studying in physical basis, yet can be solved analytically. There is an initial inventory  $N_{10}$  of  $^{239}\text{Pu}$  in a system characterized by a certain number of structural atoms  $N_S$ , having an average capture cross section  $\sigma_S$ . We fix  $N_{10}$  to be equal to unity; all molar quantities are normalized in this way. The  $^{239}\text{Pu}$  capture-to-fission ratio is  $\alpha$ . The irradiation is performed at a constant total fission rate  $= Q$ , which was held at a value of  $2.5 \text{ yr}^{-1}$ . We take  $k_{\text{eff}}$  to be a constant, maintained by the addition of a time-dependent

external feed of the  $^{239}\text{Pu}$ , which is given the name  $R_1$ . Fission products accumulate in the system, with concentration  $N_0$  and cross section  $\sigma_0$ . The cross sections change with time, representing changes in spectrum due to the concentration changes with time. Depletion of fission products due to capture is neglected. Production of actinides by capture reactions is also neglected. The governing equations for the  $^{239}\text{Pu}$  and fission product concentrations are then given by, respectively:

$$(2) \quad \frac{dN_1}{dt} = -Q(1+\alpha) + R_1$$

$$(3) \quad \frac{dN_0}{dt} = 2Q$$

Now because there is only one fissile species, the requirement of a constant  $k_{\text{eff}}$  is equivalent in this system to a constant thermal utilization  $f = 1/(1+x)$  where

$$(4) \quad x = \frac{N_s \sigma_s + N_0 \sigma_0}{N_1 \sigma_1}$$

and therefore we constrain  $x$  to be a constant. We take the cross section ratios to be functions of the concentration of the  $^{239}\text{Pu}$  and fission products in the system. We have chosen these functions to have the following form:

$$(5) \quad r_s = \frac{\sigma_s}{\sigma_1} = r_{s,0} \{ a_1(N_1 - 1) + a_0 N_0 + 1 \}$$

$$(6) \quad r_0 = \frac{\sigma_0}{\sigma_1} = r_{0,0} \{ a_1(N_1 - 1) + a_0 N_0 + 1 \}$$

Here we have chosen equations that are linear in concentration and give fixed values at beginning-of-life. The coefficients  $r_{s,0}$ ,  $r_{0,0}$ ,  $a_0$  and  $a_1$  are all positive. The values of  $r_{s,0}$ ,  $r_{0,0}$  are set equal to 0.10 in the calculations. What this means is that the structure and fission product cross sections are 10% of that of  $^{239}\text{Pu}$  at beginning of life, gradually becoming a greater fraction as time progresses. The other two coefficients are set equal to the adjustable parameter  $p$ .

We then take the derivative of  $x$  with respect to time and constrain it to be zero. From the rearrangement of this equation the feed rate of  $^{239}\text{Pu}$  is calculated:

$$(7) \quad R_1 = Q(1 + \alpha) + \frac{2Qr_0}{x} + \frac{1}{x} [N_s r_{s,0} + N_0 r_{0,0}] \left[ a_1 \frac{dN_1}{dt} + a_0 \frac{dN_0}{dt} \right]$$

Equation (7) is substituted into (2). After some rearranging we have:

$$(8) \quad \frac{dN_1}{dt} = \frac{c_3 - c_4 t}{c_1 - t} + \frac{N_1 - 1}{c_1 - t}$$

where

$$(9) \quad c_1 = \left[ 1 - \frac{N_s a_1 r_{s0}}{x} \right] / c_2$$

$$(10) \quad c_2 = \frac{2Q a_1 r_{00}}{x}$$

$$(11) \quad c_3 = \left[ \frac{2Q a_0 N_s r_{s0}}{x} + \frac{2Q r_{00}}{x} \right] / c_2$$

$$(12) \quad c_4 = \left[ \frac{8Q^2 a_0 r_{00}}{x} \right] / c_2$$

This equation (8), with the initial condition that  $N_1(t=0) = 1$ , has an exact solution:

$$(13) \quad N_1 = 1 + \frac{2c_3 t + c_4 t^2}{2(c_1 - t)}$$

We now discuss the approximate solutions. For instance, by using the binomial expansion of the denominator we can state (13) as a power series in the time variable  $t$ . The series is given below after truncation to second order:

$$(14) \quad N_1^* = 1 + \frac{c_3}{c_1} t + \left[ \frac{c_3}{c_1^2} + \frac{c_4}{2c_1} \right] t^2$$

The unmodified Euler procedure discussed above gives the following for the concentration at the end of the first time step (where  $t \rightarrow h$  from below, or  $t = h_-$ ):

$$(15) \quad N_1^u(t = h_-) = 1 + \frac{2Q r_{00} h}{x}$$

and gives the following for the concentration at the beginning of the second time interval (or  $t = h_+$ ):

$$(16) \quad N_1^u(t = h_+) = \left[ 1 + \frac{2Q r_{00} h}{x} \right] \left[ 1 + \frac{2Q r_{00} h}{x} a_1 + 2Q h a_0 \right]$$

The difference between these last two is that the newer spectral information (at  $t = h_+$ ) is included in the second calculation. Therefore whenever the spectrum is updated, there is a discontinuity in the value of the feed species. It represents an instantaneous injection of feed into the system to compensate for spectral changes.

Shown in **Figure 3** (drawn with the computer program *Mathematica*<sup>8</sup>) is a comparison of the true solution, as given by equation (13), the quadratic approximation from (14) and the unmodified Euler solution. For the purposes here, we used large values for the coefficients  $a_0$  and  $a_1$  of 0.08. The time between spectral updates is 1 year. Closest agreement occurs immediately after the spectral update. Comparison to the analytical forms (14) and (16) show that the two are in exact agreement if terms containing  $a_0^2$ ,  $a_1^2$  and  $a_0 a_1$  are neglected. That there is a visible gap between these two approximations is due to the fairly large value for  $a_0$  and  $a_1$  used in this example.

In **Figures 4 and 5** we show a comparison of the true solution with the unmodified and modified Euler method results over a larger time frame. The values of  $a_0$  and  $a_1$  used here are 0.005. Again, we have taken the time step to be twice as large for the modified method. Either method produces an error that grows with time, however the unmodified method is *always below* the true solution while the modified method gives a value that *oscillates about* the true solution. During a spectral update the value passes from below the true value to above it. More painstaking comparison shows that the average value at the spectral update is almost exactly equal to the result from the unmodified method after the spectral update. The modified method will fall completely below the true solution if the time step is too large or the computation is carried too far forward in time.

Because the unmodified method is always below the true solution while the modified method oscillates about the solution it is difficult to fairly compare the errors in the two methods. One way is to look at the effect on reaction products or decay daughters of the primary species. In this example, the species is  $^{239}\text{Pu}$ , which has a decay daughter  $^{235}\text{U}$ . The effect on the governing equations by the introduction of this species is neglected because it is present in low concentration. The decay of  $^{235}\text{U}$  itself is also neglected. Under these assumptions the  $^{235}\text{U}$  concentration is just equal to the decay constant times the area under the  $N_1$  curve. The results of this calculation for the unmodified, modified and true solution cases is given in **Figure 6**. The upper-most curve is the true curve, followed by the unmodified and modified solution results. **Figure 7** shows the effect at  $t = 8$  of a variation in the parameter  $p$ . The unmodified Euler method produces systematically larger errors although the time step is half the size.

### Coupled ORIGEN/MCNP4a Calculational Comparison

The coupling between MCNP4a and ORIGEN2.1 uses two auxiliary codes, KOLAPS and TDI. First MCNP4a computes effective one-group cross section evaluations for the 34 actinide species. These are stored as tallies in the file MCTAL. This file is read in by the code KOLAPS. This code also has its own 124-group library of fission product cross sections. There is a separate tally in MCNP4a that creates a 124-group energy spectrum, which is collapsed against the library to produce individual one-group cross sections. The fission product and actinide cross sections are then written to an ORIGEN2.1 library.

As the code ORIGEN2.1 runs it has the capacity to add enough external feed (in our examples weapon-return plutonium) in order to hold the  $k_\infty$  at some fixed value. This

updating of the concentration occurs once every internal time step, which we have fixed at 10 days. After a certain number of these internal time steps, called "recycles", the calculations are stopped and the concentrations are fed into MCNP4a in order to evaluate the cross sections again. The MCNP4a input deck is made to represent as accurately as possible the composition computed by ORIGEN2.1. This is done by the code TDI, which reads the 34 actinide species from the ORIGEN2.1 output. In addition, 62 fission product species are read from the ORIGEN2.1 output file and treated in the same way. These species, which are the ones that MCNP4a has continuous cross section evaluations for, account typically for 85% of all fission product captures in the system. The remaining 15% are compensated for by a small increase in concentration. For the very first MCNP4a input file the concentrations are read in from an auxiliary input file.

In the computational example here we have taken a homogeneous unit cell of molten salt, laden with weapon plutonium. The boundaries of the cell are reflecting, so we are actually modeling an infinite medium. The fuel salt is composed of 1/3 LiF and 2/3 BeF<sub>2</sub>. The initial loading of weapon Pu is 0.032 moles/liter and there is an initial loading of <sup>151</sup>Eu poison present at 3x10<sup>-5</sup> atom fraction. The value  $k_{\infty}$  is held fixed at 1.050. The power density in the salt is fixed at 45 W/cc.

**Figure 8** shows the results of the <sup>239</sup>Pu calculations for 1 and 2 year time steps for the modified and unmodified methods. Because we do not have an exact solution, we use the result of a modified Euler calculation with a 1/4 year between spectral updates as a benchmark. As seen before in the simplified model, there are discontinuities in the concentration of the feed species with time. Here the concentration vs. time plot is concave down because of the time-dependence of the fission product composition. This is in contrast to the simple model above where the fission products continually absorbed more neutrons without changing composition. A downward slope is also possible here. When the spectrum is updated, ORIGEN2.1 may determine that the system is above the specified  $k_{\text{eff}}$  value and will temporarily halt the injection of new feed. A negative slope is the result.

We see that the behavior here is similar to that seen in the previous example: the modified method produces solutions that oscillate about the benchmark while the unmodified method tends to lag monotonically. The departure from the true solution, which is presumably most closely approximated by the modified method with a 1/4 year time step, is minimized by the modified method with the 2 year time step compared to the unmodified method with the 1 year time step. Because there is no graphite in this system whatsoever, the system is always somewhat epithermal. The system moves farther into the resonance region after a short time. Of the molten salt systems studied, this particular case is the most radical in its departure from linearity. We have showed the early time of the system because this is where the greatest departures occur. For times greater than 5 years, the modified Euler scheme with 1 or 2 years between spectral updates or the unmodified scheme with 1 year of less between updates produces adequate results.

## Conclusions

As the fission products and higher actinides build up in a purely weapon-Pu-fueled system, a much higher inventory is required to maintain a constant  $k_{\text{eff}}$  for the system. The spectrum becomes more epithermal, and individual one-group cross sections for fissile species can vary significantly. These changes can occur rapidly, requiring very short time steps using the unmodified Euler method. The modified Euler method will give greater confidence in the results under these circumstances. In systems where there is less radical behavior, the use of the modified method may allow longer time steps. The unit cell calculations showed that the modified method with a 1 year time step between spectral updates is probably an adequate procedure for heterogeneous or homogeneous molten salt systems using reasonable power densities.

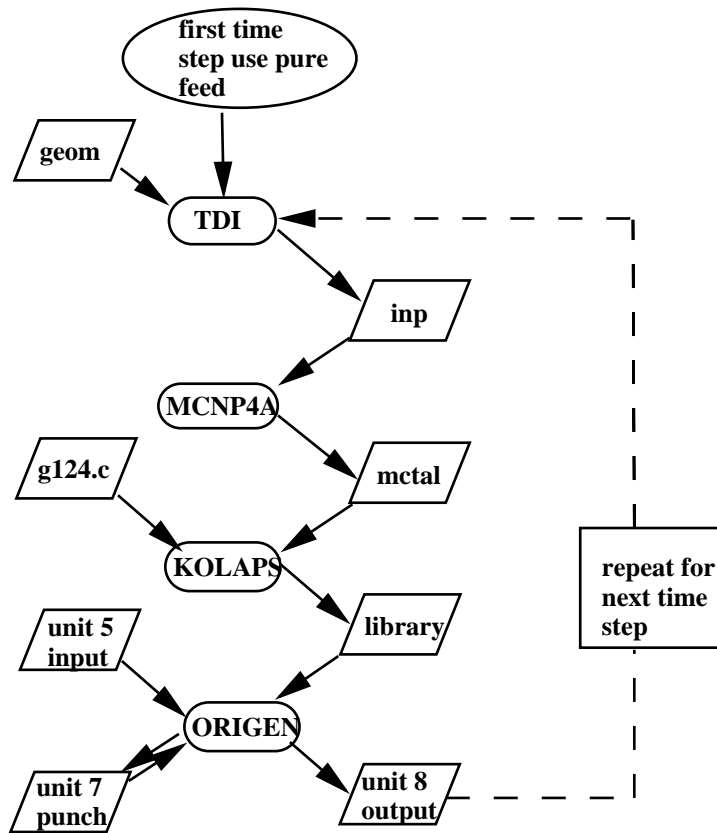


Figure 1. An unmodified Euler method for coupled transport/depletion calculations using MCNP4a and ORIGEN. Flow is controlled through a UNIX script file, which keeps track of all input and output files. Two auxiliary FORTRAN programs used are TDI and KOLAPS.

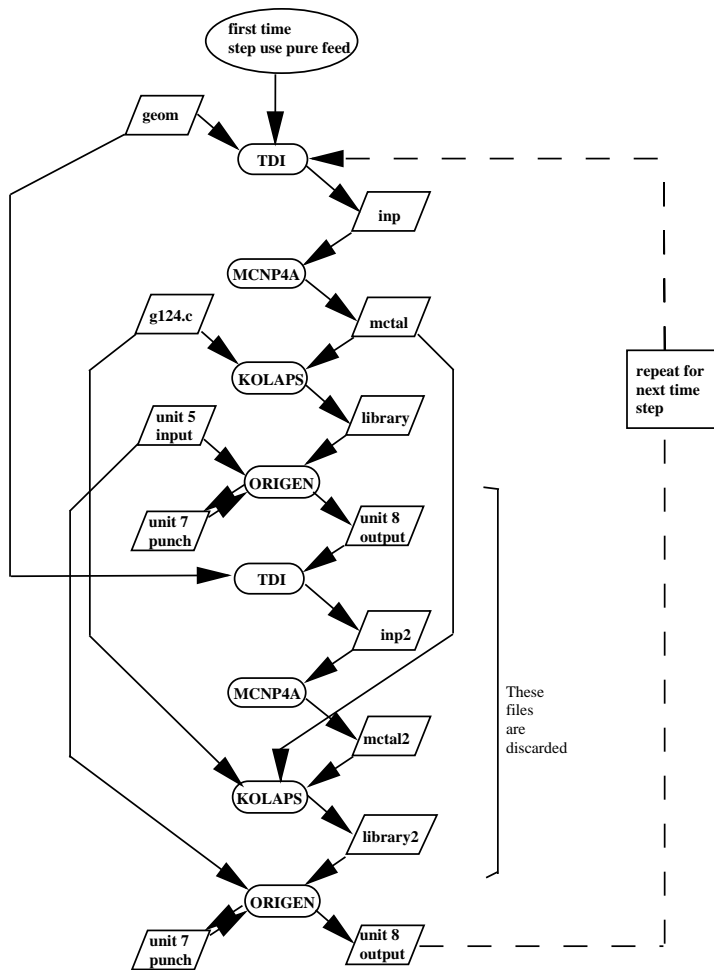


Figure 2. The Modified Euler Depletion Analysis Loop (MEDAL). The same programs are linked together in a modified Euler strategy that uses two spectral calculations per time step.

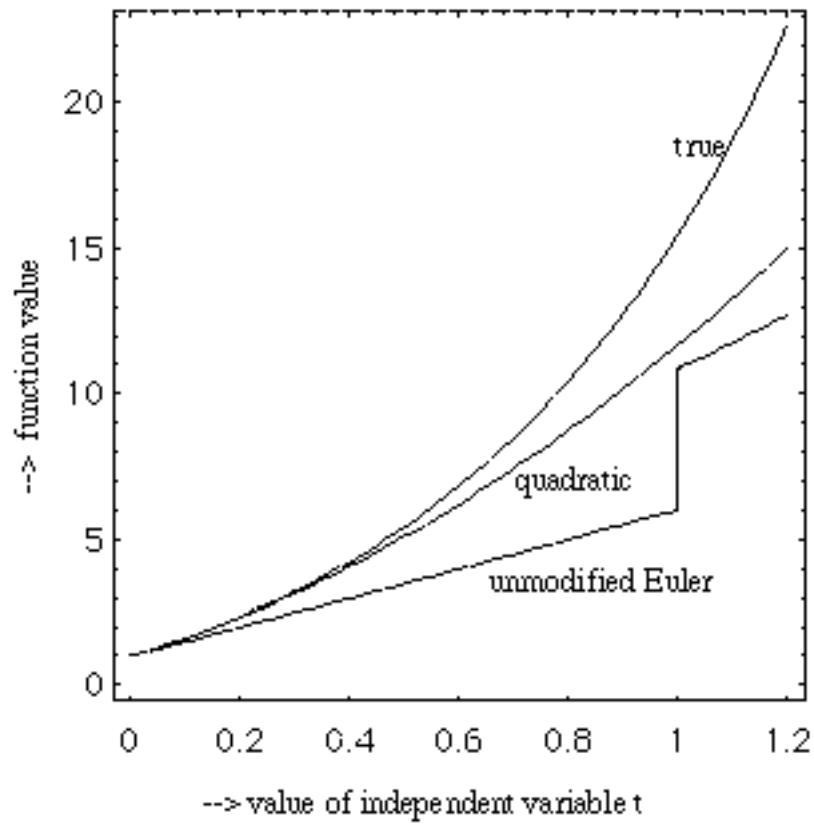


Figure 3. Comparison of the model equation results for the true solution, the quadratic approximation, and the unmodified Euler solution. All terms in the quadratic approximation except those containing the products  $a_0 \cdot a_0$ ,  $a_1 \cdot a_1$  and  $a_0 \cdot a_1$  are in the unmodified Euler solution.

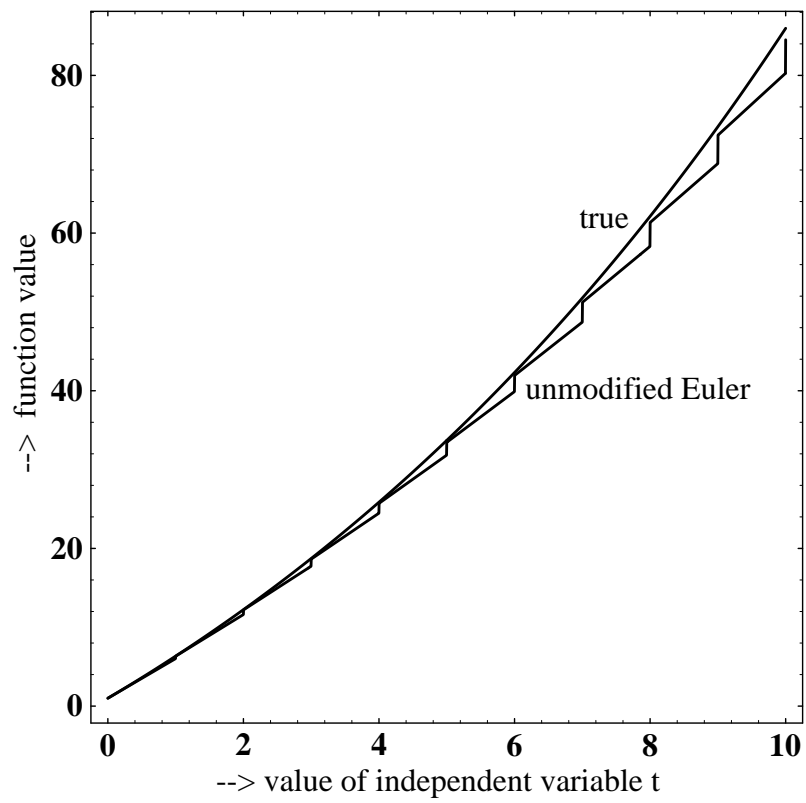


Figure 4. Comparison of the model equation results for the true solution with the unmodified Euler solution for  $p = a_0 = a_1 = 0.005$ . The approximation always stays below the true solution.

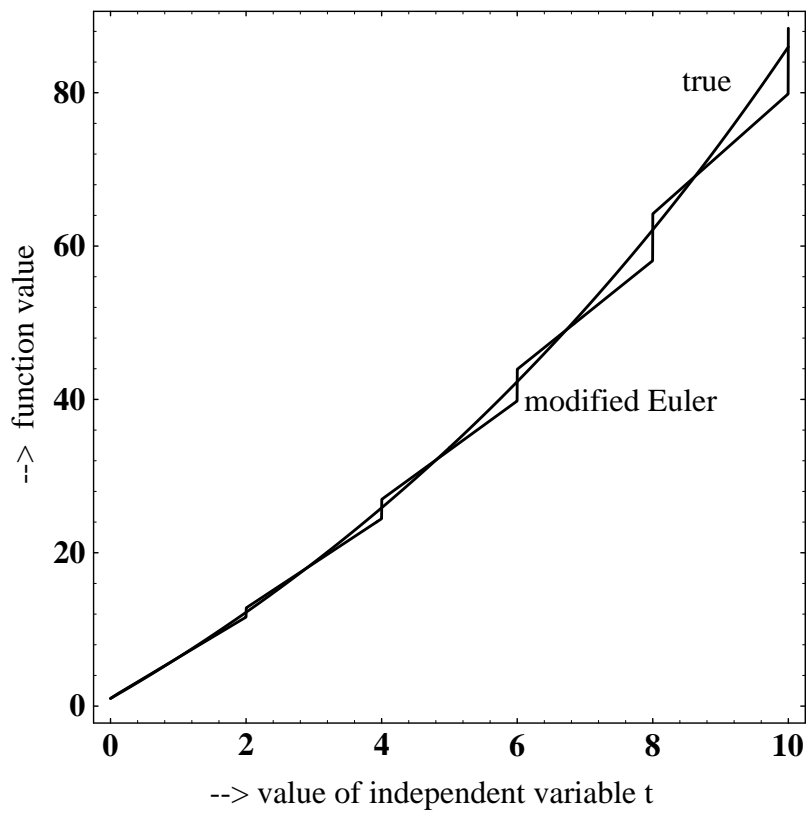


Figure 5. Comparison of the model equation results for the true solution with the MEDAL solution for  $p = a_0 = a_1 = 0.005$ , using double the time-step. The agreement on average is much closer.

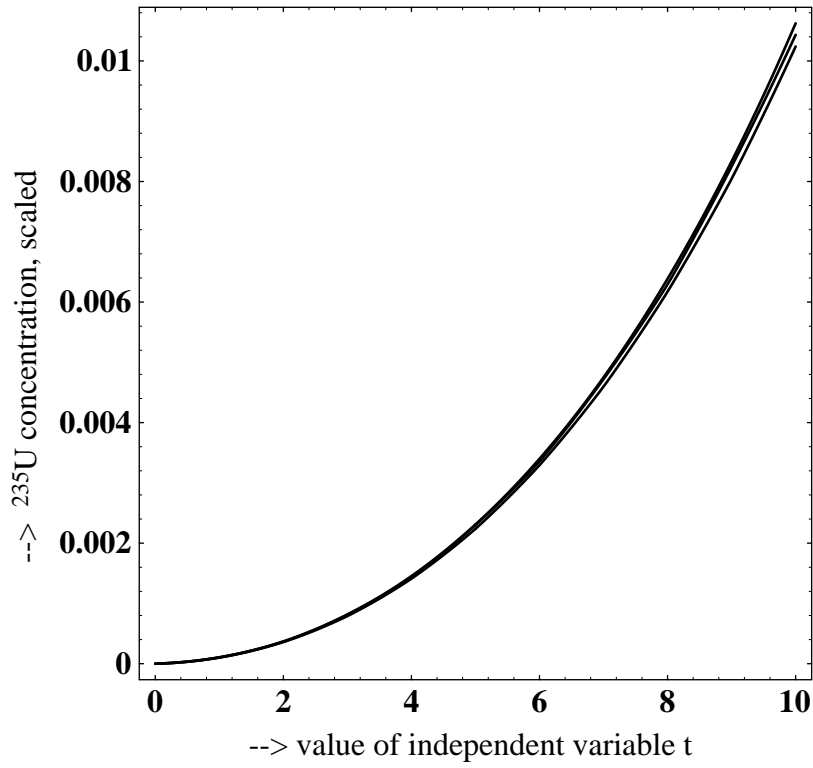


Figure 6. Comparison of the modified and unmodified methods for the U-235 daughter product of Pu-239 versus time for  $p = a_0 = a_1 = 0.005$ . We have used the area underneath a Pu-239 calculation to compute this graph. The upper curve is the true curve, followed by the modified and the unmodified approximations.

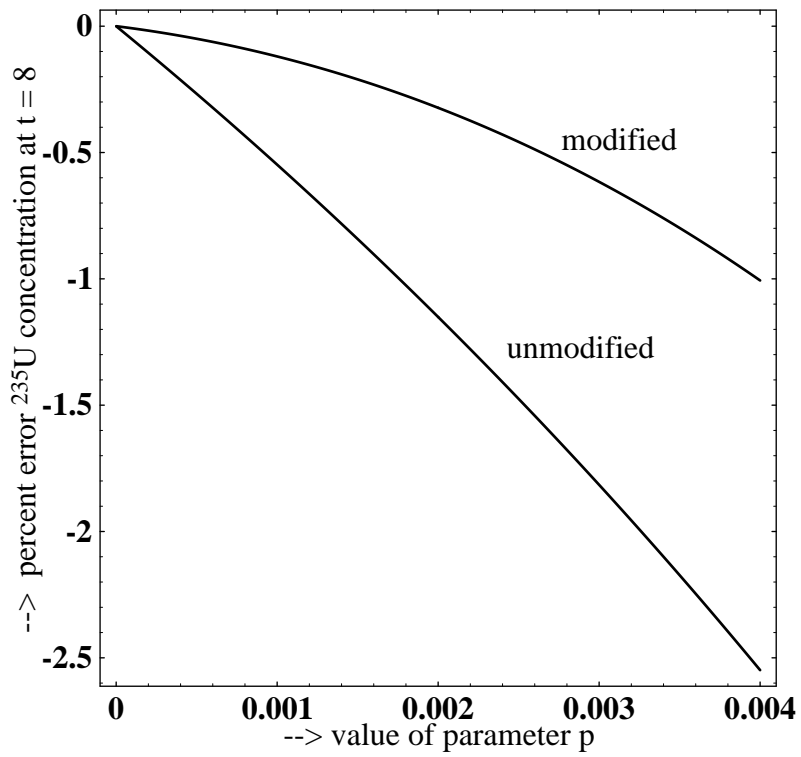


Figure 7. Comparison of the modified and unmodified methods for the U-235 decay product of Pu-239 versus  $p = a_0 = a_1$  at a fixed  $t = 8$ . As was seen in Figure 4, the shape of the curve near  $p = 0$  is different in the two cases.

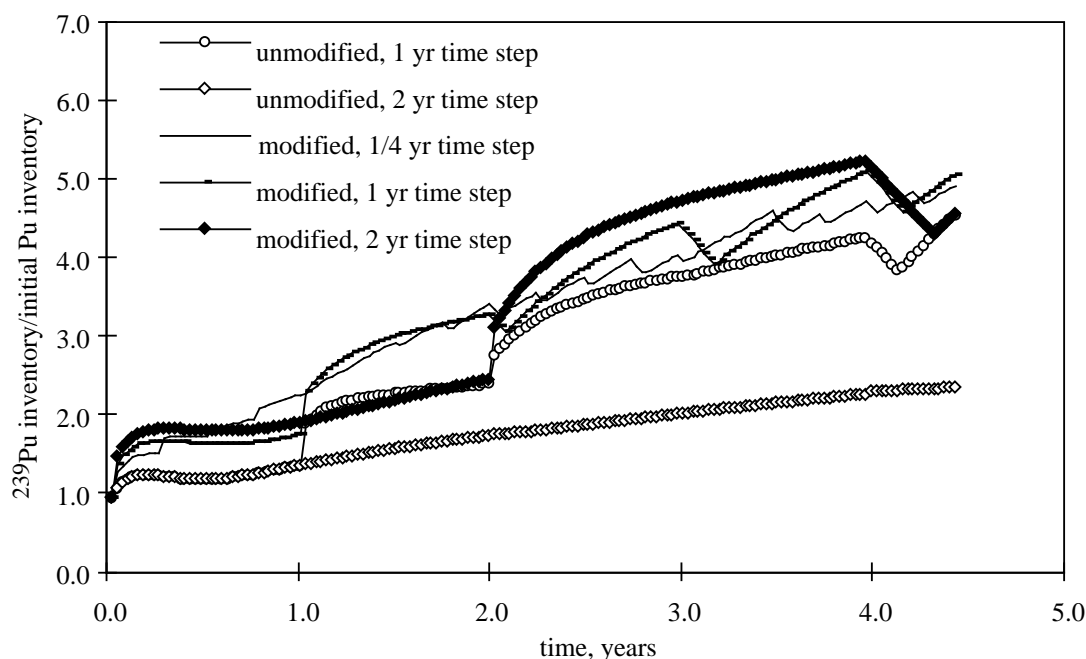


Figure 8. Time dependence of the  $^{239}\text{Pu}$  inventory in a homogeneous unit cell calculation using the modified and unmodified Euler methods to couple MCNP4 and ORIGEN2.1 together. Because we lack an exact solution in this case, we have taken a "modified" calculation with 3 months between spectral updates as a benchmark for comparison. Similar behavior to the analytical model is seen, where the unmodified solution lags the benchmark, the modified method oscillates about the benchmark.

<sup>1</sup>Croff, A. G., ORIGEN2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials, *Nucl. Technol.* **62** pg. 335, 1983.

<sup>2</sup>Kochurov, B. P., "An Advanced Method of Heterogeneous Reactor Theory", JAERI-Review 94-002.

<sup>3</sup>Breismeister, J. F., *MCNP-A General Monte Carlo N-Particle Transport Code*, LA-12625-M, Los Alamos National Laboratory, Los Alamos, NM, 1993.

<sup>4</sup>D. Garber (Editor), "ENDF/B-V", Report BNL-17541 (ENDF-201) National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY, (October 1975).

<sup>5</sup>R. J. Howerton, et. al., "The LLL Evaluated Nuclear Data Library (ENDL): Evaluations Techniques, Reaction Index and Descriptions of Individual Reactions", Lawrence Livermore National Laboratory report UCRL-50400, Vol. 15, Part A (September 1975).

<sup>6</sup>R. W. Hamming, "Numerical Methods for Scientists and Engineers, Second Edition", McGraw Hill Book Co. New York, 1973.

<sup>7</sup>R. S. Babcock, D. E. Wessol, C. A. Wemple, and S. C. Mason, "The MOCUP Interface: A Coupled Monte Carlo/ Depletion System", Proceedings of the 1994 Topical Meeting on Advances in Reactor Physics, Knoxville, TN April 11-15, 1994, American Nuclear Society, LaGrange Park, IL.

<sup>8</sup>Wolfram, S., "Mathematica, a system for doing mathematics by computer, Second Ed." Advanced Book Program, Addison-Wesley Publishing Co. Redwood City, CA, 1991.